

Non-Monotonic Temperature Dependence of Coulomb Drag Peaks in Graphene

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Coulomb drag is a direct measurement of the electron-electron interactions between two electronic layers. Graphene is a versatile electronic material with a high-degree of tunability opening up regimes that were not previously accessible. All previous theoretical studies of graphene Coulomb drag away from charge neutrality assume a spatially homogeneous carrier density which gives a peak in the Coulomb drag that decreases with temperature in contradiction to available experimental results. In this work, we develop an effective medium theory for Coulomb drag and show that including spatial inhomogeneity in the carrier density gives rise to a non-monotonic temperature dependence of the drag peaks that is in quantitative agreement with experimental data. Our results also show that at double-charge neutrality, there is a large negative momentum drag for correlated density fluctuations that competes with energy drag and is also non-monotonic with temperature. In addition, we show that when the density fluctuations in the two layers are correlated, the disordered theory has less symmetry than the homogeneous case, giving rise to a violation of Onsager reciprocity between the active and passive layers.

The effects of electron-electron interactions in transport measurements are usually a small correction to predictions from models of non-interacting electrons. Coulomb drag is special because it is identically zero unless interactions are present, making it an ideal experimental probe of electron-electron interactions. A typical experiment measuring drag involves driving a current in one (active) layer and measuring the induced potential drop caused by the Coulomb force in a physically separated (passive) layer. The electric field accompanying this potential drop is then easily deduced and divided by the current density in the active layer to yield the drag resistivity. Studies of this effect now have a history of almost thirty years. The first experiments [1–3] were performed using double layer two-dimensional electronic gases (i.e. GaAlAs heterostructures), followed by a series of associated theoretical works [4–8]. Studying drag is also interesting from an applications viewpoint. Under certain conditions, strong Coulomb coupling between electron and hole layers causes the formation of stable excitons that can Bose-Einstein condense to a superfluid ground state with the potential for application in low power electronics (see Ref. [9] and references therein). Coulomb drag measurements are the standard method for verifying the presence of such a condensate which shows strong signatures in the drag resistivity [10–12].

Graphene’s high level of electronic tunability makes it an ideal platform to study Coulomb drag [13] and exciton condensation [14, 15]. At high carrier density, experimental studies of drag in graphene reproduced observations previously made in GaAs, but close to charge neutrality, graphene probed new regimes that were not previously accessible experimentally [16–18]. Graphene Coulomb drag experiments continue to reveal new physics. Just this year, drag experiments between two bilayer graphene

sheets suggested at new mechanisms of Coulomb drag beyond the usual ones of energy and momentum [19, 20] and observed a quantized Hall drag resistivity [21, 22] that is consistent with condensation of excitons across the two sheets. Contemporaneous with the first experimental studies of graphene Coulomb drag were several theoretical works (e.g. Refs. [23–29]), from which the following picture emerged: Away from charge neutrality, the carrier density is assumed to be spatially homogeneous and the drag resistivity ρ_d is universal (in that it does not depend on the in-plane scattering mechanism or disorder concentration) and dominated by momentum exchange between the layers. At charge neutrality, the transport is assumed to be ballistic, the carrier density is assumed to be spatially inhomogeneous and drag resistivity is dominated by energy exchange mediated by the existence of regions of correlated carrier density fluctuations about zero average density.

The theory for momentum drag with equal density in the active and passive layers has ρ_d vanish both at zero carrier density and large carrier density with a peak in drag resistivity ρ_d^{peak} that is independent of any impurity details and that decreases exponentially with increasing temperature occurring at an intermediate carrier density $n_{\text{peak}} \sim (k_B T / \hbar v_F)^2$ where v_F is graphene Fermi velocity. That this peak in drag resistivity decreases with temperature T , is in contradiction to all published experimental data. Surprisingly, this obvious discrepancy was not noticed previously in the literature.

In this work we focus on the question of momentum drag in two graphene monolayers separated by a thin dielectric spacer in an experimental configuration such as described in Ref. [17]. We directly address the discrepancy between the theoretical predictions of a drag resistivity peak that decreases with temperature and the

experimental observation to the contrary. The experimental single-sheet mobilities of $\sim 100,000 \text{ cm}^2/\text{Vs}$ suggests [30] diffusive transport in the layers (i.e. mean-free path much smaller than sample size) and a carrier density fluctuation $n_{\text{rms}} \sim 5 \times 10^{10} \text{ cm}^{-2}$. Since $n_{\text{rms}} \sim n_{\text{peak}}$ for most of the experimental temperature regime, a spatially homogeneous theory of momentum drag is inadequate. Typically, effective medium theories (EMT) have been used to treat the problem of spatially inhomogeneous carrier densities or ‘charge puddles’ in single layer systems. These theories work by considering a network of patches with conductivities $\sigma(r)$ embedded in an effective homogeneous medium with conductivity σ_E and requir-

ing that the spatially integrated electric field of patches with $\sigma(r) \neq \sigma_E$ vanishes. In our generalization of EMT to the case of Coulomb drag, we impose the same requirement in each of the two layers while simultaneously taking into account the small electric field corrections within each patch due to the interlayer Coulomb collisions. Finally, we assume that the interlayer interaction does not change the passive layer conductivity. This is justified by the fact that drag conductivities are typically very small compared to the in-plane conductivity of either layer. This procedure leads to our main theoretical result, which is the first development of an effective medium theory applicable for Coulomb drag. We find:

$$\sigma_D^E = \frac{\int_{-\infty}^{\infty} dn'_A \int_{-\infty}^{\infty} dn'_P P(n'_A, n'_P, \eta) \cdot \left[\frac{\sigma_D(n'_A, n'_P) \sigma_A^E}{(\sigma_A^E + \sigma_A(n'_A))(\sigma_P^E + \sigma_P(n'_P))} \right]}{\int_{-\infty}^{\infty} dn'_A \int_{-\infty}^{\infty} dn'_P P(n'_A, n'_P, \eta) \cdot \left[\frac{\sigma_A(n'_A)}{(\sigma_A^E + \sigma_A(n'_A))(\sigma_P^E + \sigma_P(n'_P))} \right]}, \quad (1)$$

where n'_A and n'_P denote the charge densities in the active and passive layers and σ_D^E is the effective medium theory averaged drag conductivity obtained by solving the equation. A detailed derivation of Eq. (1) together with the details of how various terms in it are computed can be found in the Supplemental Material [31]. Equation (1) contains the following inputs: $\sigma_D(n_A, n_P)$ is the homogeneous drag conductivity between two sheets with uniform densities n_A and n_P , and σ_A^E and σ_P^E are the effective medium theory averaged monolayer conductivities readily obtained by solving [32],

$$\int_{-\infty}^{\infty} dn_i P(n_i) \frac{\sigma(n_i) - \sigma_i^E}{\sigma(n_i) + \sigma_i^E} = 0, \quad (2)$$

where $i = A, P$ denotes the active and passive layer index and $\sigma(n_i)$ is the homogeneous conductivity of a graphene sheet with uniform density n_i . The double layer distribution $P(n_A, n_P, \eta)$ is defined as the joint probability of a randomly chosen point on the active layer having a local charge density n_A with the point on the passive layer directly above having density n_P . We model this by the bivariate normal probability distribution which involves three parameters – n_{rms}^A (n_{rms}^P) is the root mean square density fluctuations in the active (passive) layer and $-1 \leq \eta \leq 1$ parameterizes the degree of correlation between the two layers [31]. The monolayer distributions $P(n_i)$ appearing in Eq. (2), use the usual (monovariate) Gaussian probability function obtained from the bivariate distribution by integrating out one of the layer densities. In the limit of two perfectly correlated puddles of equal area fraction, our EMT result agrees with previous calculation [33].

We first analyze in some detail the existing drag theory (i.e. homogeneous with no puddles present) for σ_D

before moving on to the inhomogeneous case since the homogeneous case is used as an input for our EMT in Eq. (1). This also allows us to highlight the inadequacy of the existing theory for explaining experimental data. The homogeneous drag physics is well-described in several previous works [13, 24, 26–28] but our discussion here nonetheless contains new results beyond what is available in the literature. The drag conductivity $\sigma_D(n_A, n_P)$ between two homogeneous graphene sheets is given by

$$\sigma_D = \frac{1}{16\pi k_B T} \int_{-\infty}^{\infty} \frac{d^2 q}{(2\pi)^2} \int_{-\infty}^{\infty} \frac{d\omega}{\sinh^2(\frac{\hbar\omega}{2k_B T})} \Gamma^x \left(\omega, \mathbf{q}, \frac{\mu_A}{k_B T} \right) \times \Gamma^x \left(\omega, \mathbf{q}, \frac{\mu_P}{k_B T} \right) |V(q, \omega, d)|^2, \quad (3)$$

where T is temperature, μ_i is chemical potential of the i -th layer, d is interlayer spacer width, $V(q, \omega, d)$ is dynamically screened interlayer Coulomb interaction, and Γ^x refers to the x -component of the nonlinear susceptibility in monolayer graphene [31]. While Eq. (3) in its general form is common in the literature, we differ from previous works in the details for treating the non-linear susceptibility in that we use a realistic energy-dependent scattering time. This causes an unphysical logarithmic divergence in σ_D [26], which we fix by making use of the full dynamical polarizability function at finite temperature, e.g. as given in Ref. [34]. Most previous treatments use an unrealistic energy independent scattering time to circumvent this problem. However, calculating drag resistivity with an energy-dependent scattering time makes a quantitative difference (within a factor of 2) compared with assuming an energy-independent scattering time [31]. In Eq. (3), σ_D is written as a function of the layer chemical potentials μ rather than charge densities n

although we often refer to σ_D as a function of the latter. This is because at fixed temperature, there is a one-to-one correspondence between charge density and chemical potential [31], making the two terms interchangeable.

Figures 1(a) and (b) show the Coulomb drag σ_D as a function of chemical potential (left panel) and carrier density (right panel) respectively. In all our figures, we assume either identically doped layers $\mu_A = \mu_P \equiv \mu$ and $n_A = n_P \equiv n$, or $\mu_A = -\mu_P \equiv \mu$ and $n_A = -n_P \equiv n$ since these are the typical situations studied in experiments. The physics of σ_D involves three factors – the strength of screening, the volume of phase space available for scattering, and the electron-hole asymmetry within each layer. Screening does two things: It screens electrons in each layer from the surrounding impurities and also screens the electrons in the two layers from each other. The former (latter) effect tends to increase (decrease) σ_D . Which of the two effects dominate is determined only by doing the actual calculation. A larger amount of scattering phase space tends to make σ_D larger since more inter-layer collisions can take place. Finally, a greater degree of electron-hole asymmetry favours larger σ_D , because in the absence of such asymmetry, identical electron and hole currents will cancel each exactly, yielding zero drag current and thus vanishing σ_D . These physical mechanisms and their competition provide a qualitative description of the non-monotonic dependence of $\sigma_D(\mu/k_B T)$ shown in Fig. 1(a). At $\mu/k_B T = 0$, $\sigma_D = 0$ for all T because of electron-hole symmetry. As $\mu/k_B T$ increases at each fixed T , electron-hole asymmetry increases, leading to larger σ_D . At very large $\mu/k_B T$, strong screening between the layers begins to effectively decouple them, leading to decreasing σ_D . Increasing T at fixed $\mu/k_B T$ (i.e. fixed electron-hole asymmetry) leads to increasing σ_D due to the increase in scattering phase space. Thus we have a good qualitative understanding of $\sigma_D(\mu/k_B T)$ computed numerically in Fig. 1(a).

In addition to the numerical result, we can also do analytical perturbative expansions of Eq. (3). There are three energy scales associated with drag conductivity, namely temperature $k_B T$, the effective interlayer energy exchange cutoff introduced by the spacer $\hbar v_F/d$, and the chemical potential μ of the layers, where we assume for simplicity that $\mu_A = \mu_P \equiv \mu$. There are thus six different limits defined by the various possible relative magnitudes of these energies. In recent experiments [17, 18], d is on the order of nanometers while T ranges from 50 – 300 K. Therefore, $k_B T d/\hbar v_F \lesssim 0.1$ and we are left with only 3 experimentally relevant limits to consider: (i) low density $\mu \ll k_B T \ll \hbar v_F/d$, (ii) intermediate density $k_B T \ll \mu \ll \hbar v_F/d$ and (iii) high density $k_B T \ll \hbar v_F/d \ll \mu$. We can extract semi-analytic expressions in all three limits.

In both the low and intermediate density limits, σ_D may be approximated by expanding it to lowest non-

vanishing order in $\mu/k_B T$ giving

$$\sigma_D = \frac{e^2}{h} \frac{1}{2r_s^2} \left(\frac{\mu}{k_B T} \right)^2 \frac{(k_B T)^4}{(\hbar v_F)^4 n_{\text{imp}}^2} F \left(r_s, \frac{k_B T d}{\hbar v_F} \right), \quad (4)$$

where the function F is of the form

$$F(r_s, x) = a e^{-bx} + c, \quad (5)$$

with a, b and c being numerically determined constants. For experimentally relevant $r_s = 0.57$, $a = 0.46, b = 10.25, c = 0.04$ (see Supplemental Material [31] for other values of r_s). This result is plotted as dashed lines in Fig. 1(a) and shows good agreement with the full numerical solution for small values of $\mu/k_B T$. We note that this result has not been reported previously in the literature.

In the high density limit, the drag conductivity is given by [13, 26–28]

$$\sigma_D = \frac{e^2}{h} \frac{\zeta(3)}{32\pi} \left(\frac{1}{J[2r_s] n_{\text{imp}} d^2} \right)^2 \left(\frac{\mu}{k_B T} \right)^{-2}, \quad (6)$$

where

$$\frac{J[x]}{x^3} = \frac{\pi}{4} + 3x - \frac{3\pi x^2}{2} + \frac{x(3x^2 - 2) \arccos(1/x)}{\sqrt{x^2 - 1}}. \quad (7)$$

The $J[x]$ expression was first calculated by one of the authors in the context of single layer graphene [30]. For $r_s = 0.57$, $J[2r_s] \approx 0.1$. This result is plotted in dash-dotted lines in Fig. 1(a). Notice that this high-density asymptote does not match the full numerical curves even at $\mu/k_B T$ as large as 25, showing the limited utility of this expansion for comparison with experimental data.

In Fig. 1(b) we replot drag conductivity as a function of carrier density. This also results in a non-monotonic curve that vanishes for both low and high carrier density. However, some subtle features emerge. In the high density limit, increasing T increases σ_D due to increased scattering phase-space. The opposite happens at the low density limit (see inset) where increasing T leads to a drop in σ_D as we are then approaching the limit of perfect electron-hole symmetry where σ_D vanishes. We emphasize that although the physical mechanisms are the same, σ_D curves can look very different and show different temperature dependence depending on whether it is plotted as a function of $\mu/k_B T$ or n . We can understand the subtleties by calculating corresponding analytical results for $\sigma_D(n)$. For instance, in the low density regime $\mu \ll k_B T \ll \hbar v_F/d$, $\mu = E_F^2/(4 \log(2) k_B T)$ and one obtains

$$\sigma_D = \frac{e^2}{h} \frac{1}{r_s^2} \left(\frac{n}{n_{\text{imp}}} \right)^2 \frac{\pi^2}{32(\log 2)^2} F \left(r_s, \frac{k_B T d}{\hbar v_F} \right). \quad (8)$$

This result implies that the drag conductivity decreases with temperature at very low densities and our analytics agree with the full numerics (see the inset of Fig. 1(b),

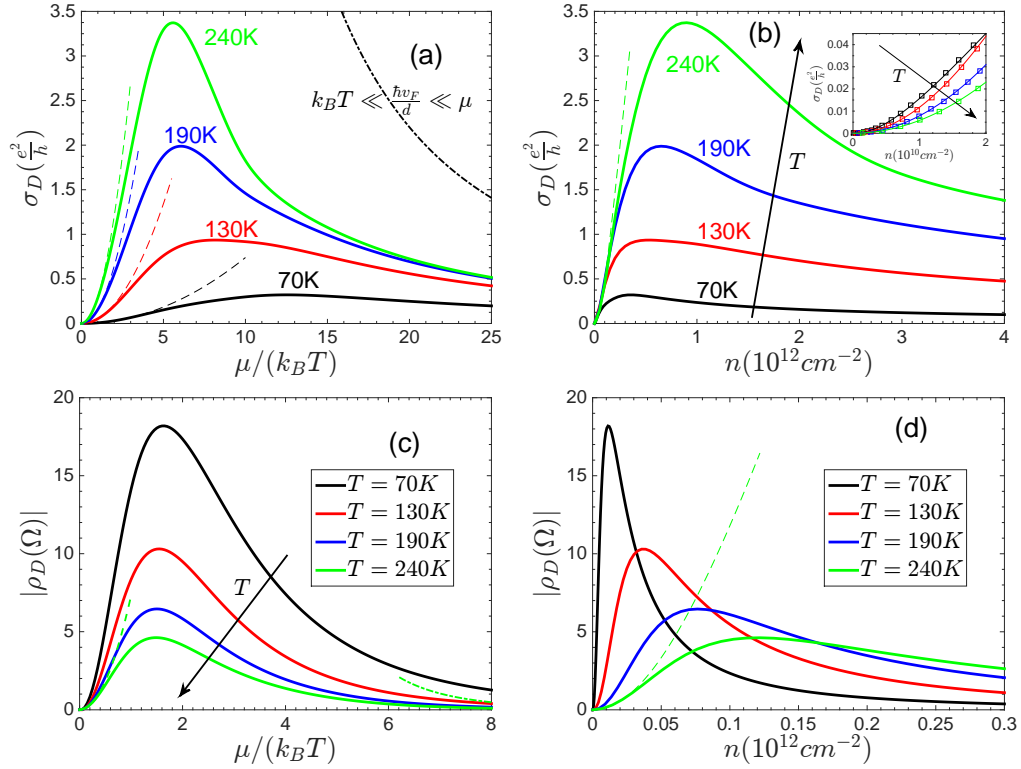


Figure 1. (color online) Coulomb Drag theory with homogeneous carrier density. Upper panels show drag conductivity σ_D at $T = 70, 130, 190, 240$ K as a function of (a) chemical potential $\mu/k_B T$ and (b) carrier density n . Low (high) density asymptotes are plotted in dashed (dash-dotted) lines. The inset of (b) is a zoom-in at low density and we represent our asymptotes (numerical data points) using lines (squares). In homogeneous theory, σ_D is an even function of both μ and n . Lower panels show drag resistivity ρ_D at the same temperatures as a function of (c) $\mu/k_B T$ and (d) n . In all panels, impurity density $n_{\text{imp}} = 5 \times 10^{10} \text{ cm}^{-2}$, spacer distance $d = 9 \text{ nm}$ and identically doped layers $\mu_A = \mu_P \equiv \mu$ and $n_A = n_P \equiv n$ are assumed. For neatness, we display the low density asymptote for only one temperature in panels (b)-(d).

where we have zoomed in on the very low density part of the curves). In the intermediate density limit $k_B T \ll \mu \ll \hbar v_F/d$, $\mu = E_F$ in Eq. (4) and we obtain

$$\sigma_D = \frac{e^2}{h} \frac{1}{2r_s^2} \frac{n}{n_{\text{imp}}} \frac{\pi(k_B T)^2}{(\hbar v_F)^2 n_{\text{imp}}} F\left(r_s, \frac{k_B T d}{\hbar v_F}\right), \quad (9)$$

showing that σ_D at fixed n now increases with T with the T^2 prefactor dominating over the weak T -dependence of $F(r_s, k_B T d/\hbar v_F)$ in this regime. Both our analytics and numerics show a clear crossover in temperature dependence as we go from low to intermediate density. These low and intermediate density asymptotes mentioned above have not been reported previously in the literature.

Next, we consider the drag resistivity ρ_D , the quantity typically measured in drag experiments, given by

$$\rho_D \approx -\frac{\sigma_D}{\sigma_A \sigma_P}. \quad (10)$$

The drag resistivity depends not just on the drag conductivity but also monolayer conductivities. The physics of ρ_D depends on the interplay of the same three factors

as σ_D , however unlike σ_D , it depends not on the average speed at which charges move in the passive layer, but rather the amount of charge that has accumulated at the boundaries after the system has reached equilibrium. Because ρ_D depends on the monolayer conductivities, this makes the interpretation more complicated than that for σ_D . We plot drag resistivity as a function of $\mu/k_B T$ and n in Figs. 1(c) and (d). Contrary to what one might naively expect, increasing temperature (at fixed $\mu/k_B T$) always decreases ρ_D . This is because the monolayer conductivities σ_A and σ_P have a stronger temperature dependence which is well-known in the literature [31]. However, as a function of carrier density $\rho_D(n)$, an interesting crossover occurs in the temperature dependence of ρ_D . For low n and most of the experimental regime, ρ_D decreases with T because we are approaching perfect electron-hole symmetry. But at large n , ρ_D increases with T [13, 26–28] because we are deep in the limit of strong screening and high electron-hole asymmetry and the main effect of increasing T is to increase the phase space available for interlayer energy exchange. We also compute the low (dashed lines) and high density (dot-dashed) asymptotes,

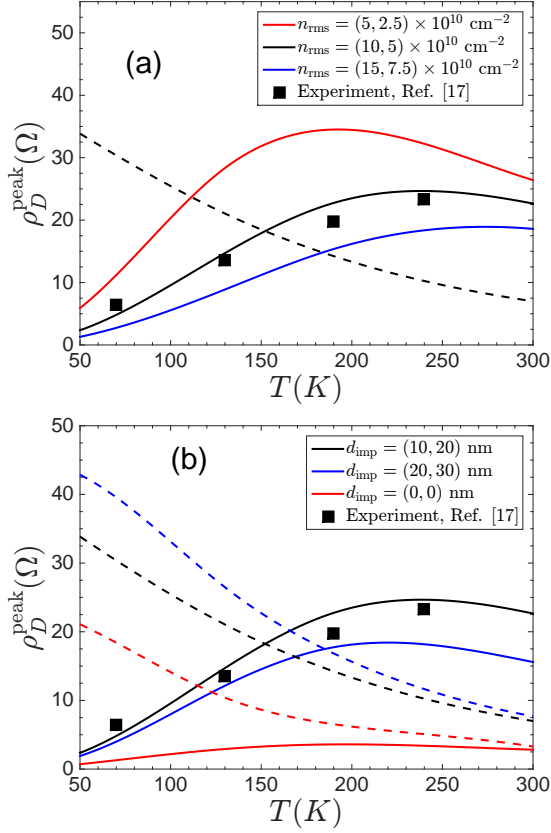


Figure 2. (color online) Effective medium theory results for Coulomb drag. (a) The finite density peak drag resistivity ρ_D^{peak} along the $n_A = -n_P = n$ line as a function of temperature for different inhomogeneity strengths. $d_{\text{imp}} = (10, 20) \text{ nm}$. Dashed lines represent numerical calculations of ρ_D^{peak} in the absence of puddles which show the opposite behaviour compared to experiment. (b) The same as panel (a) but for different values of d_{imp} . Here, $n_{\text{rms}} = (8, 4) \times 10^{10} \text{ cm}^{-2}$. The dashed lines again represent numerics for ρ_D^{peak} without puddles. Experimental data points from Ref. [17] represented by black squares and interlayer spacing $d = 9 \text{ nm}$ and $\eta = 0.5$ in both panels.

obtained by combining the appropriate drag and mono-layer conductivity asymptotes in Eq. (10) and confirm their consistency with the numerics. For $\rho_D(n)$, we omit the high density asymptote (which just like that of $\sigma_D(n)$ in Fig. 1(a) is very inaccurate in the range of densities shown).

As seen in Fig. 1(d), $|\rho_D(n)|$ is more complicated than $|\rho_D(\mu/k_B T)|$ just as $\sigma_D(n)$ was more complicated than $\sigma_D(\mu/k_B T)$. $|\rho_D(n)|$ peaks at some finite density and then decreases. The temperature dependence of these peaks is of particular interest. At each temperature, ρ_D reaches its peak when $\mu \approx k_B T$. We obtain an estimate of $|\rho_D^{\text{peak}}|$ as a function of T by setting $\mu/k_B T = 1$ in our expressions for σ_D and $\sigma_{A,P}$ and find that for the homogeneous theory, the value of $|\rho_D^{\text{peak}}|$ decreases monotonically with

T as

$$|\rho_D^{\text{peak}}| \sim \exp\left(-10.25 \frac{k_B T d}{\hbar v_F}\right). \quad (11)$$

This result (verified numerically) is contrary to the experimentally measured peaks (at finite density) of Ref. [17] (plotted in Fig. 2) that clearly increase with T . This is a significant contradiction between theory and experiment that requires an explanation.

We address this discrepancy by calculating the drag peaks in the presence of electron-hole puddles using the EMT discussed earlier. The EMT inputs to Eq. (10) are obtained by solving Eqs. (1) and (2) with the homogeneous theory for σ_D , σ_A and σ_P as inputs. Our results, shown in Fig. 2, are in good quantitative agreement with the experimental data. In the calculation, we use $\eta = 0.5$ but note that this value does not significantly change the value of ρ_D^{peak} . The puddles cause the drag peaks to behave non-monotonically as a function of T , first increasing at low temperatures and reverting back to the homogeneous (i.e. monotonically decreasing) behaviour of Eq. (11) at high temperatures.

We can understand this because the EMT roughly averages ρ_D over a density interval of size $\sim n_{\text{rms}}$. At low temperatures when $\hbar v_F \sqrt{\pi n_{\text{rms}}} \gtrsim k_B T$, this causes the high density (i.e. $\mu \gg k_B T$) temperature dependence of ρ_D (i.e. increasing with T) to move to lower densities where the drag peak occurs. At very high temperatures where $\hbar v_F \sqrt{\pi n_{\text{rms}}} \ll k_B T$, this averaging does not affect the T -dependence because the energy window over which averaging takes place is much smaller than $k_B T$ and the regimes $\mu \gg k_B T$ and $\mu \ll k_B T$ are not mixed in the averaging, allowing the homogeneous temperature dependence at any n to be relatively unaffected by the averaging process. In Fig. 2(a), we show how the non-monotonicity depends on the sample purity $n_{\text{rms}} \equiv (n_{\text{rms}}^A, n_{\text{rms}}^P)$, while in (b) we discuss how it depends on distance of the impurity plane from the electronic sheets $d_{\text{imp}} \equiv (d_{\text{imp}}^A, d_{\text{imp}}^P)$. The higher density fluctuation strength in one layer reflects the experimental set-up where one layer is nearer to the underlying SiO_2 substrate where charged impurities reside [17, 35]. Increasing d_{imp} serves to increase the screening between carriers and charged impurities, while keeping the screening between the layers unchanged, so we expect drag resistivity to increase monotonically with d_{imp} and this is verified in the homogeneous theory calculation shown in dashed lines of Fig. 2(b). In the presence of puddles, the drag peaks develop a non-monotonic dependence on d_{imp} .

We emphasize that our EMT establishes that the fact that experiments see drag peaks increasing with temperature is explained by the presence of puddles. Our results also allow us to predict that if the drag experiments of Refs. [17, 18] are done at a slightly higher temperature, the drag peaks observed will show a non-monotonic be-

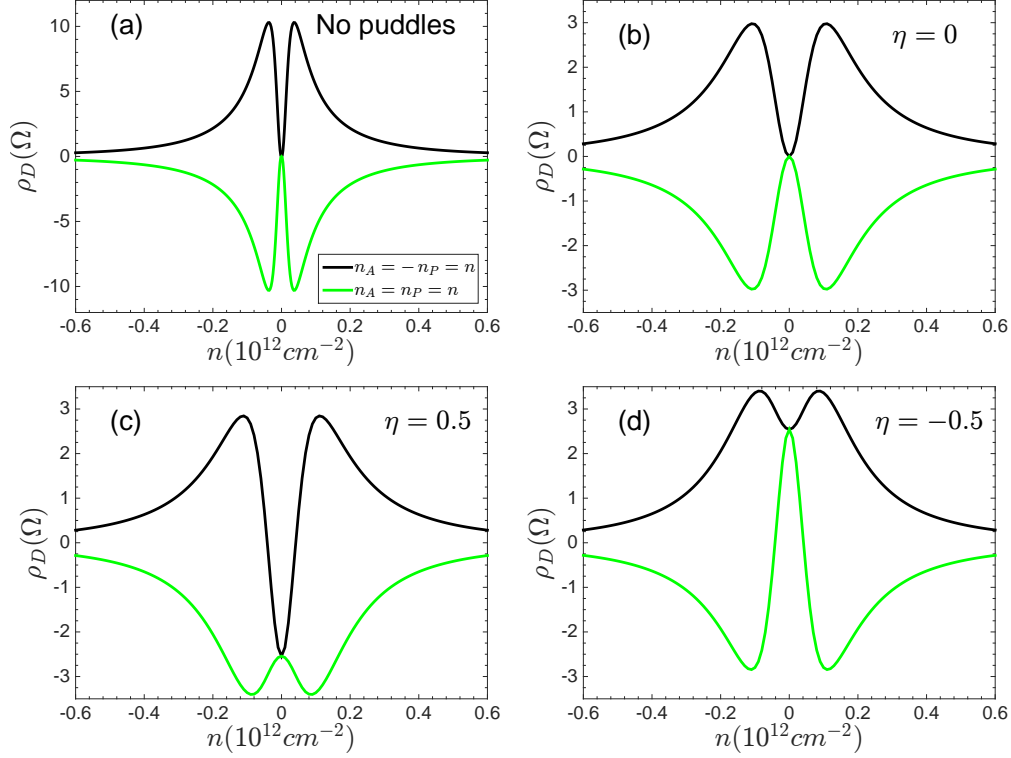


Figure 3. (color online) The role of layer correlations on drag density. ρ_D calculated as a function of charge density along the lines of oppositely matched densities $n_A = -n_P = n$ (upper curve) and identically matched density $n_A = n_P = n$ (bottom curve) in the case of (a) perfect charge homogeneity (b) uncorrelated puddles $\eta = 0$ (c) correlated puddles $\eta = 0.5$ and (d) anti-correlated puddles $\eta = -0.5$. We assume $d_{\text{imp}}^A = d_{\text{imp}}^P = 0$, $T = 130$ K and interlayer spacing $d = 9$ nm. In panels (b)-(d), $(n_{\text{rms}}^A, n_{\text{rms}}^P) = (10, 5) \times 10^{10} \text{ cm}^{-2}$. The ρ_D^{CN} is an odd function of η .

haviour with respect to temperature. Such an observation will be an important test of the theory developed here.

We now address the role of correlated puddles on ρ_D as a function of density, including at the double charge neutrality point ρ_D^{CN} . For simplicity we set $d_{\text{imp}} = (0, 0)$. Fig. 3(a) shows (as a benchmark) the homogeneous calculation without puddles. The black (upper) curve considers electron-hole layers (i.e. $n_A = -n_P = n$) and the red (lower) curve shows double layers of the same carrier type ($n_A = n_P = n$). Panel (b) includes uncorrelated puddles ($\eta = 0$). Notice that the features are qualitatively similar to the homogeneous case (panel (a)), but with broader peaks smeared by n_{rms} . In particular, even with inhomogeneous carrier density, without interlayer puddle correlation, $\rho_D = 0$ at double charge neutrality. Panel (c) shows the effect of correlated disorder ($\eta = 0.5$), and in this case the symmetry of $\rho_D(n_A, -n_P) = -\rho_D(n_A, n_P)$ is broken. Moreover, at double charge neutrality, there is a negative Coulomb drag (i.e. $\rho_D^{\text{CN}} = \rho_D(n = 0) < 0$). Panel (d) shows that for anti-correlated puddles, there is a positive Coulomb drag at the double charge neutrality and $\rho_D^{\text{CN}}(-\eta) = -\rho_D^{\text{CN}}(\eta)$. We note that $\rho_D^{\text{CN}}(T)$ is non-monotonic as shown in Fig. 4 because each pair of

puddles can be viewed as a tiny region of homogeneous drag layers in which drag behaves non-monotonically as explained earlier, and averaging over many such pairs preserves the non-monotonic dependence. In contrast, the energy-drag invoked by Song and Levitov [29] to explain the data of Ref. [17] at double charge neutrality has the opposite sign of ρ_D^{CN} compared to the momentum drag we discuss here. Our work thus shows that these two mechanisms always compete. In the presence of correlated (or anti-correlated) puddles, either mechanism can potentially dominate depending on parameters. We note that at double charge neutrality both mechanisms show a T^2 dependence at low temperature and a T^{-4} dependence at high temperature regimes even though they differ in sign. However, the momentum drag discussed here should always dominate away from double charge neutrality and is responsible for the ‘outer’ (i.e. finite density) peak ρ_D^{peak} observed experimentally.

Notice that in panels (c) and (d), there is a slight difference in the size of the ‘outer’ peaks ρ_D^{peak} depending on whether one measures along the line of identical or opposite signs of densities e.g. the magnitude of the black peaks in Fig. 3(c) ($\eta = 0.5$) is larger than the magnitude

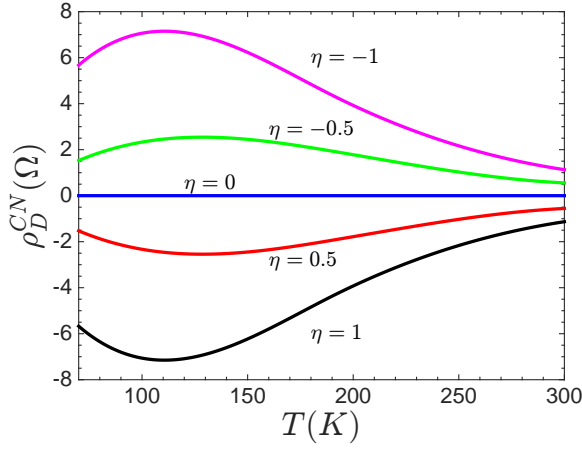


Figure 4. (color online) Non-monotonic temperature dependence of drag at double charge neutrality. The drag resistivity ρ_D^{CN} at the Dirac point as a function of temperature for different interlayer correlation coefficients η . $(n_{\text{rms}}^{\text{A}}, n_{\text{rms}}^{\text{P}}) = (10, 5) \times 10^{10} \text{ cm}^{-2}$. Interlayer spacing $d = 9 \text{ nm}$. Finite correlation between layers is necessary for non-zero drag resistivity at charge neutrality.

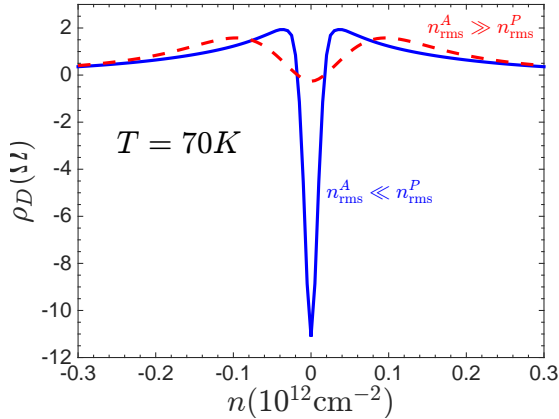


Figure 5. (color online) Apparent violation of Onsager relation in the effective medium theory. ρ_D as a function of charge density $n_{\text{A}} = n_{\text{P}} = n$ for $n_{\text{rms}}^{\text{A}} = 1 \times 10^{10} \text{ cm}^{-2}$, $n_{\text{rms}}^{\text{P}} = 10 \times 10^{10} \text{ cm}^{-2}$ (blue solid) and for swapped layers $n_{\text{rms}}^{\text{A}} = 10 \times 10^{10} \text{ cm}^{-2}$, $n_{\text{rms}}^{\text{P}} = 1 \times 10^{10} \text{ cm}^{-2}$ (red dashed). In both cases, correlation coefficient $\eta = 0.5$. Interlayer spacing $d = 9 \text{ nm}$.

of the green dip in the same panel. Since energy drag is negligible away from the Dirac point, this suggests a way to experimentally determine the nature of the interlayer correlations (i.e. correlated, anti-correlated or uncorrelated) by comparing the peaks along these two lines in density space. Although these differences are small, they could provide a definitive way to resolve this controversy in the experimental literature. This correlation-induced asymmetry is much larger away from the high symmetry

cuts $n_{\text{A}} = \pm n_{\text{P}}$ [31].

Finally, we note that our Coulomb drag EMT result Eq. (1) is not symmetric between the active and passive layers, implying that violations of Onsager's reciprocity relation are possible. In the context of drag, Onsager's reciprocity [36] predicts that there should be no difference in ρ_D when the active and passive layers are switched. We have found that if there exist nonzero correlations (i.e. $\eta \neq 0$) between the layers, violations will occur in the majority of cases [31]. Experimental observation of Onsager reciprocity violation in drag systems has been reported in Refs. [16], [37] and [38].

In summary, we have developed an effective medium theory for Coulomb drag in graphene to handle the carrier density fluctuations that are known to be important in graphene experiments. Our formalism resolves an important but previously ignored discrepancy where the Coulomb drag peaks increase (decrease) with temperature in experiment (theory). We predict that the temperature dependence of ρ_D^{peak} is non-monotonic, increasing at low temperature but decreasing at higher temperature as $\exp\left(-\frac{k_B T d}{\hbar v_F}\right)$. We also discuss the drag resistivity at double charge neutrality and show that the momentum drag and energy drag compete giving contributions of opposite sign. Moreover, the apparent violation of Onsager's relation observed in recent experiments is naturally explained by our formalism. Finally, we suggest comparing data for the peaks along $n_{\text{A}} = n_{\text{P}}$ and $n_{\text{A}} = -n_{\text{P}}$ to determine whether the puddles in graphene double layers are correlated or anti-correlated, thereby resolving an ongoing controversy in the literature.

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SUPPLEMENTAL MATERIAL FOR NON-MONOTONIC DEPENDENCE OF COULOMB DRAG IN GRAPHENE

DERIVATION OF EQ. (1)

We consider the standard drag setup – two parallel 2D sheets of identical size separated by some finite distance, with one layer being the passive and the other the active. Each layer is made up of N patches/puddles of differing conductivities, σ_i^A and σ_i^P , where $i = 1, \dots, N$. We shall assume that the puddles in both layers are circles of radius a and lie exactly atop each other, with the i th puddle of the active layer lying exactly atop the i th puddle of the passive, as in Fig. S1. We do not make any further assumptions about the nature of these puddles in our formulation of the EMT and our derivation is applicable regardless of whether the puddles are correlated, anti-correlated or uncorrelated, etc.

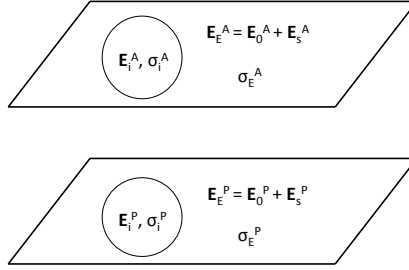


Figure S1. The i th puddles of the active and passive layers are each embedded inside their own effective media.

Unlike the single layer case detailed in the next section, there are now three effective conductivities to be determined. These are the effective in-plane conductivities for the active and passive layers, and the effective drag conductivity between the layers. They are denoted by σ_E^A, σ_E^P and σ_E^D respectively. Our final result will be the three equations (see Eqs. (1) and (2)) whose solutions are these effective conductivities.

We briefly summarize the steps of our derivation before delving into the details. First, we take the i th pair of puddles (where i is any of the N patches), one from each layer, and embed each one inside its own homogeneous effective medium of conductivity σ_E^A and σ_E^P respectively as shown in Fig. S1, where each effective medium has within it the same uniform effective field (excluding the field caused by the puddle), denoted by \vec{E}_0^P and \vec{E}_0^A respectively and the drag conductivity between the two effective media is denoted σ_E^D . Next, we determine the electric fields inside the puddles, \vec{E}_i^A and \vec{E}_i^P . Finally, we substitute these into the EMT self-consistency equations, given by

$$\sum_i f_i \vec{E}_i^A = \vec{E}_0^A, \quad (\text{S1})$$

and

$$\sum_i f_i \vec{E}_i^P = \vec{E}_0^P \quad (\text{S2})$$

where f_i refers to the areal fraction of the i th patch relative to the whole layer, and obtain Eqs. (1) and (2). Note that $\sum_i f_i = 1$.

We now begin the detailed derivation. Our first task is to solve for the field inside the i th (where i is arbitrary) puddle that has been embedded in the effective medium as described above. The potentials and fields within each layer and puddle are of the same form as Eqs. (S24) to (S29). That is, we assume that the puddles are regions of uniform

2D polarization in a direction parallel to the effective electric field of the layer. These polarizations are denoted by $\vec{M}^A = M^A \vec{e}_x$ and $\vec{M}^P = M^P \vec{e}_x$ for the active and passive layers respectively. In the active effective medium, we have

$$U_E^A(r, \theta) = -E_0^A r \cos(\theta) + \frac{a^2}{2\epsilon_0} \frac{M^A}{r} \cos(\theta) \quad (S3)$$

and

$$\vec{E}_E^A(r, \theta) = E_0^A \vec{e}_x + \frac{a^2}{2\epsilon_0} \frac{1}{r^2} \left[2(\vec{M}^A \cdot \vec{e}_r) \vec{e}_r - \vec{M}^A \right]. \quad (S4)$$

Note that we have chosen a radial coordinate system with its origin at the center of the two puddles. Inside the i th puddle of the active effective medium, we guess (as we do for the single layer case) that the field is simply proportional to the effective field

$$U_i^A(r, \theta) = -C^A E_0^A r \cos(\theta) \quad (S5)$$

and

$$\vec{E}_i^A(r, \theta) = C^A E_0^A \vec{e}_x, \quad (S6)$$

where C^A is an unknown constant to be determined. The uniqueness theorem guarantees that any successful (i.e. it obeys all boundary conditions) guess corresponds to the actual physical solution. The exact same considerations apply for the passive layer. That is, the previous four equations with all ‘A’ superscripts replaced by ‘P’s describe the passive layer.

We thus have a total of four unknowns, M^A , M^P , C^A and C^P . These are solved for by making use of the following boundary conditions. First, the potentials must be continuous at the boundaries of the puddle in each layer. That is,

$$U_E^A(r = a, \theta) = U_i^A(r = a, \theta) \quad (S7)$$

and

$$U_E^P(r = a, \theta) = U_i^P(r = a, \theta) \quad (S8)$$

for all θ , with the explicit forms of the potentials as given in the previous paragraph. Second, the radial current density must also be continuous at these boundaries. The current densities in the active and passive effective media \vec{j}_E^A and \vec{j}_E^P are related to the electric fields \vec{E}_E^A and \vec{E}_E^P by the matrix equation

$$\begin{pmatrix} \vec{j}_E^A \\ 0 \end{pmatrix} = \begin{pmatrix} \sigma_E^A & \sigma_E^D \\ \sigma_E^D & \sigma_E^P \end{pmatrix} \begin{pmatrix} \vec{E}_E^A \\ \vec{E}_E^P \end{pmatrix}, \quad (S9)$$

where $\vec{j}_E^P = 0$ because we consider the situation in which the passive layer is in an open-circuit configuration. Note that \vec{E}_E^A points in the direction of the driving current in the active layer. Within the puddles, we have the similar relation

$$\begin{pmatrix} \vec{j}_i^A \\ 0 \end{pmatrix} = \begin{pmatrix} \sigma_i^A & \sigma_i^D \\ \sigma_i^D & \sigma_i^P \end{pmatrix} \begin{pmatrix} \vec{E}_i^A \\ \vec{E}_i^P \end{pmatrix}. \quad (S10)$$

The electric fields \vec{E}_E^A and \vec{E}_E^P are either parallel or anti-parallel on physical grounds, since the latter is caused by the former through the drag effect. We choose our axes so that both fields are along the x -axis. Explicitly, $\vec{E}_E^A = E_E^A \vec{e}_x$ and $\vec{E}_E^P = E_E^P \vec{e}_x$. Equations (S9) and (S10) together with the requirement of continuous radial current density yield

$$\sigma_E^A E_{E,r}^A + \sigma_E^D E_{E,r}^P = \sigma_i^A E_{i,r}^A + \sigma_i^D E_{i,r}^P \quad (S11)$$

and

$$\sigma_E^D E_{E,r}^A + \sigma_E^P E_{E,r}^P = \sigma_i^D E_{i,r}^A + \sigma_i^P E_{i,r}^P, \quad (S12)$$

where the subscript r denotes the radial component. Substituting the potentials and fields in Eqs. (S3) to (S6) and their counterparts for the passive layer into Eqs. (S7), (S8), (S11) and (S12) yields four equations for the four

unknowns mentioned. Note that the θ dependence drops out of the problem, leaving behind only the magnitudes of the various vectors defined. We solve for C^A and C^P via the usual methods of simultaneous equations to obtain

$$C^A = \left(1 - \frac{2(\sigma_i^D \sigma_E^P - \sigma_E^D \sigma_i^P) \frac{E_0^P}{E_0^A} + (\sigma_i^A - \sigma_E^A)(\sigma_E^P + \sigma_i^P) - ((\sigma_i^D)^2 - (\sigma_E^D)^2)}{(\sigma_E^A + \sigma_i^A)(\sigma_E^P + \sigma_i^P) - (\sigma_E^D + \sigma_i^D)^2} \right) \quad (S13)$$

and

$$C^P = \left(1 - \frac{2(\sigma_i^D \sigma_E^A - \sigma_E^D \sigma_i^A) \frac{E_0^A}{E_0^P} + (\sigma_i^P - \sigma_E^P)(\sigma_E^A + \sigma_i^A) - ((\sigma_i^D)^2 - (\sigma_E^D)^2)}{(\sigma_E^A + \sigma_i^A)(\sigma_E^P + \sigma_i^P) - (\sigma_E^D + \sigma_i^D)^2} \right). \quad (S14)$$

We have thus solved for the electric fields within the i th pair of puddles. In order for these fields to be combined with the self-consistency equations in a useful manner however, we must first be able to write the electric field inside each puddle as a function of only its own layer's effective medium field (i.e., instead of being a function of the effective medium fields of both layers). We achieve this by requiring that given the two effective medium layers without any puddles (i.e., all the puddles have already been effectively taken into account by the effective medium fields), there will be no current flow in the passive layer. The expression for this condition is given by

$$\frac{E_0^A}{E_0^P} = -\frac{\sigma_E^P}{\sigma_E^D}. \quad (S15)$$

Substituting Eqs. (S13) and (S14) into Eqs. (S6) and its passive layer counterpart respectively and making use of Eq. (S15), we obtain the intra-puddle field in the active (passive) layer as a function of only the active (passive) layer's effective medium electric field. Explicitly, we obtain for the active layer puddle

$$E_i^A = \left(1 - \frac{2(\sigma_E^D \sigma_i^P - \sigma_i^D \sigma_E^P) \frac{\sigma_E^D}{\sigma_E^P} + (\sigma_i^A - \sigma_E^A)(\sigma_E^P + \sigma_i^P) - ((\sigma_i^D)^2 - (\sigma_E^D)^2)}{(\sigma_E^A + \sigma_i^A)(\sigma_E^P + \sigma_i^P) - (\sigma_E^D + \sigma_i^D)^2} \right) E_0^A \quad (S16)$$

and

$$E_i^P = \left(1 - \frac{2(\sigma_E^D \sigma_i^A - \sigma_i^D \sigma_E^A) \frac{\sigma_E^P}{\sigma_E^D} + (\sigma_i^P - \sigma_E^P)(\sigma_E^A + \sigma_i^A) - ((\sigma_i^D)^2 - (\sigma_E^D)^2)}{(\sigma_E^A + \sigma_i^A)(\sigma_E^P + \sigma_i^P) - (\sigma_E^D + \sigma_i^D)^2} \right) E_0^P \quad (S17)$$

for the passive layer puddle.

Finally, we substitute these into the self-consistency equations (S1) and (S2) and make the approximation of setting all terms quadratic in drag conductivities to zero since drag conductivities are typically much smaller (six orders of magnitude less) than in-plane conductivities. This yields

$$\sum_i f_i \cdot \frac{\sigma_i^A - \sigma_E^A}{\sigma_E^A + \sigma_i^A} = 0. \quad (S18)$$

and

$$\sum_i f_i \cdot \frac{2(\sigma_E^D \sigma_i^A - \sigma_i^D \sigma_E^A) \frac{\sigma_E^P}{\sigma_E^D}}{(\sigma_E^A + \sigma_i^A)(\sigma_E^P + \sigma_i^P)} + \sum_i f_i \cdot \frac{\sigma_i^P - \sigma_E^P}{\sigma_E^P + \sigma_i^P} = 0, \quad (S19)$$

where we have made use of the fact that $\sum_i f_i = 1$. Equation (S18) is the already-known discrete single layer EMT equation applied to the active layer. Equation (S19) is more complicated and comprises two terms summing to zero. Since the two unknowns σ_E^D and σ_E^P cannot be determined by this single equation, we require another condition. Since the drag conductivity is very small compared to the in-plane conductivity within either layer, we may obtain this condition by approximating that the interlayer interaction has a negligible effect on the passive layer conductivity. This then implies that the single layer EMT equation (cf. (S18)) applies to the passive layer, and the two terms of Eq. (S19) must both be individually zero. Equating the first term with zero yields

$$\sum_i f_i \cdot \frac{2(\sigma_E^D \sigma_i^A - \sigma_i^D \sigma_E^A) \frac{\sigma_E^P}{\sigma_E^D}}{(\sigma_E^A + \sigma_i^A)(\sigma_E^P + \sigma_i^P)} = 0, \quad (S20)$$

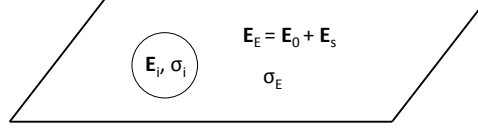


Figure S2. A circular region of inhomogeneity embedded inside a homogeneous effective medium of conductivity σ_E .

while doing the same to the second term yields

$$\sum_i f_i \cdot \frac{\sigma_i^p - \sigma_E^p}{\sigma_E^p + \sigma_i^p} = 0. \quad (\text{S21})$$

The former is a newly derived discrete EMT drag equation while the latter is just the already-known discrete single layer EMT equation applied to the passive layer. Note that in the case of there being only two puddles in each layer (relevant only at double charge neutrality) so that $f_i = \frac{1}{2}$, $i = 1, 2$, we recover from the above expressions the results of Ref. [33]. Generalizing the above discrete EMT equations to the continuum, we obtain Eqs. (1) and (2).

To model the continuous distribution of puddles, we employ the bivariate normal probability distribution

$$P(n'_A, n'_P, \eta) = \frac{1}{2\pi n_{\text{rms}}^A n_{\text{rms}}^P \sqrt{1 - \eta^2}} \exp \left(-\frac{1}{2(1 - \eta^2)} \left[\frac{(n'_A - n_A)^2}{(n_{\text{rms}}^A)^2} + \frac{(n'_P - n_P)^2}{(n_{\text{rms}}^P)^2} - \frac{2\eta(n'_A - n_A)(n'_P - n_P)}{n_{\text{rms}}^A n_{\text{rms}}^P} \right] \right), \quad (\text{S22})$$

where n_{rms}^A and n_{rms}^P represent the root mean square density fluctuations of the two layers about their mean densities n_A and n_P , and the interlayer correlation coefficient η quantifies the degree to which the charge density fluctuations in the two layers are correlated or anti-correlated. The dummy integration variables n'_A and n'_P are distinguished from the average densities n_A and n_P with prime superscripts. η is defined mathematically as

$$\eta \equiv \frac{\langle (n'_A - n_A)(n'_P - n_P) \rangle}{n_{\text{rms}}^A n_{\text{rms}}^P}, \quad (\text{S23})$$

where the angular brackets refer to averaging over the density distribution of the two layers. A value of $\eta = 1$ (-1) corresponds to perfectly correlated (anti-correlated) charge density fluctuations within the two layers. We note that there are claims in the literature that the charge puddles are correlated [29] (anti-correlated [17]) due to static charged impurities (ripples on the graphene sheets). Both of these scenarios are easily modeled in Eq. (S22) by choosing the value of η accordingly.

DERIVATION OF EQ. (2)

For completeness, we also review the derivation of the monolayer EMT result, Eq. (2). Useful reviews of monolayer EMT may be found in Refs. [39] and [40]. Consider a sheet of 2D material which is made up of a patchwork of N areas (i.e. puddles) each with differing conductivities σ_i , where $i = 1, \dots, N$. The areal fraction of the i th puddle is denoted by f_i with $\sum_i f_i = 1$. We wish to calculate the effective conductivity of this sheet. We first imagine that each puddle is embedded in a homogeneous effective medium of conductivity σ_E and through which permeates a uniform electric field \vec{E}_0 . Next, we work out the electric field \vec{E}_i inside each puddle in the form $\vec{E}_i = (\dots)\vec{E}_0$. Consider the i th puddle embedded in the effective medium as shown in Fig. S2. For simplicity, we assume that all puddles are circles of radius a . We find the electric field inside this puddle.

The conductivity of the medium is σ_E and it is permeated by a uniform field $\vec{E}_0 = E_0 \vec{e}_x$ corresponding to an external field that we will refer to as the primary field. We denote the field inside the puddle as \vec{E}_i and the field outside as \vec{E}_E . We assume that the puddle possesses a uniform polarization \vec{M} pointing in the same direction as the external

field, so that $\vec{M} = M\vec{e}_x$. From electrostatics, such a circular puddle is associated with an electric field in the region outside it given by

$$E_s(\vec{r}) = \frac{a^2}{2\epsilon_0 r^2} \left[2(\vec{M} \cdot \vec{e}_r) \vec{e}_r - \vec{M} \right] \quad (\text{S24})$$

with a corresponding potential

$$U_s(\vec{r}) = \frac{a^2}{2\epsilon_0} \frac{M}{r} \cos(\theta). \quad (\text{S25})$$

Here, the subscript ‘s’ is short for ‘secondary’ and we placed the origin of our radial coordinate system at the center of the circular puddle. Hence, the total field outside ($r > a$) is, taking the \vec{e}_r components,

$$\begin{aligned} E_{E,r}(r, \theta) &= E_{0,r} + E_{s,r} \\ &= E_0 \cos(\theta) + \frac{a^2}{2\epsilon_0 r^2} M \cos(\theta) \end{aligned} \quad (\text{S26})$$

and the potential is

$$U_E(r, \theta) = -E_0 r \cos(\theta) + \frac{a^2}{2\epsilon_0} \frac{M}{r} \cos(\theta). \quad (\text{S27})$$

Next, we consider the field inside the puddle. We guess that the field inside is proportional to the externally applied field \vec{E}_0 . Hence,

$$\begin{aligned} \vec{E}_i &= C \vec{E}_0 \\ &= C E_0 \vec{e}_x \\ &= C E_0 \cos(\theta) \vec{e}_r \end{aligned} \quad (\text{S28})$$

with potential

$$U_i(r, \theta) = -C E_0 r \cos(\theta). \quad (\text{S29})$$

We now use boundary conditions to solve for the unknown C , and obtain the desired field inside the puddle. The boundary conditions that must be obeyed at the boundary of the puddle are the continuity of potential, and continuity of radial current density. In equations, they are

$$U_E(a, \theta) = U_i(a, \theta) \quad (\text{S30})$$

and

$$\sigma_E E_{E,r} = \sigma_i E_{i,r}. \quad (\text{S31})$$

We can make use of these two boundary conditions to solve for the two unknowns M and C . This yields

$$C = \frac{2\sigma_E}{\sigma_E + \sigma_i} \quad (\text{S32})$$

and

$$M = 2\epsilon_0 E_0 \left(\frac{2\sigma_E}{\sigma_E + \sigma_i} - 1 \right). \quad (\text{S33})$$

The fact that solutions for C and M exist validates our earlier guess in Eq. (S29), which we know is the unique physical solution due to the uniqueness theorem. We are not interested in M here. Our objective was to find \vec{E}_i , which we have successfully done by determining C . Explicitly, we have found that

$$E_i = \left(\frac{2\sigma_E}{\sigma_E + \sigma_i} \right) E_0, \quad (\text{S34})$$

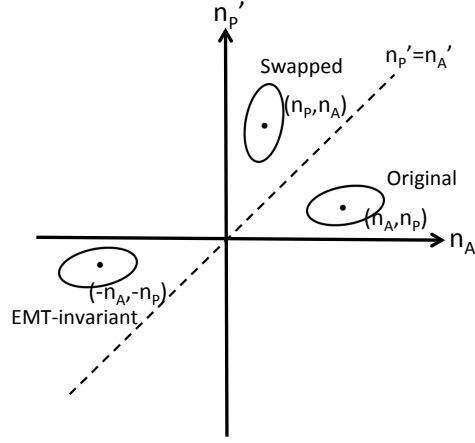


Figure S3. A graphical aid for checking for Onsager violation. If the ‘original’ oval corresponding to the initial drag conductivity before the swapping of layers is mapped onto the ‘EMT-invariant’ oval or left unchanged by the swap, then Onsager reciprocity will be obeyed, provided the two layers also have identical density fluctuation amplitudes.

where E_i is the magnitude of \vec{E}_i . We substitute the above into the EMT self-consistency condition

$$\sum_{i=1}^N f_i \vec{E}_i = \vec{E}_0, \quad (\text{S35})$$

and simplify to obtain

$$\sum_{i=1}^N f_i \cdot \frac{\sigma_i - \sigma_E}{\sigma_i + \sigma_E} = 0. \quad (\text{S36})$$

Generalizing to a continuum of puddles and denoting the continuous puddle label by n , one obtains Eq. (2).

S1. Symmetries of EMT drag and Violation of Onsager Reciprocity

We elaborate upon the various scenarios where violations of Onsager reciprocity occur. We first show that if the puddles in the layers are uncorrelated, or if one of the layers is homogeneous, there can be no violation of Onsager reciprocity. Lastly, we give a general graphical method (see Fig. S3) of predicting when violations will occur.

If $\eta = 0$ in Eq. (1), we may rewrite the joint probability $P(n'_A, n'_P, \eta = 0)$ as a simple product of two Gaussian distributions $P(n'_A)P(n'_P)$ and separate the integral in the denominator into two one-dimensional integrals. Then, making use of Eq. (2), we obtain

$$\sigma_D^E = \frac{\int_{-\infty}^{\infty} dn'_A \int_{-\infty}^{\infty} dn'_P P(n'_A) P(n'_P) \cdot \frac{\sigma_D(n'_A, n'_P)}{(\sigma_A^E + \sigma_A(n'_A))(\sigma_P^E + \sigma_P(n'_P))}}{\int_{-\infty}^{\infty} dn'_A \frac{P(n'_A)}{(\sigma_A^E + \sigma_A(n'_A))} \cdot \int_{-\infty}^{\infty} dn'_P \frac{P(n'_P)}{(\sigma_P^E + \sigma_P(n'_P))}}. \quad (\text{S37})$$

Recalling that $\sigma_D(x, y) = \sigma_D(y, x)$, this expression is manifestly symmetric in the two layers and proves that Onsager’s reciprocity relation will be obeyed by drag layers possessing uncorrelated charge inhomogeneities.

It is also easy to show that if one of the root-mean-density fluctuations n_{rms} goes to zero (i.e. if one layer is homogeneous), the drag conductivity is symmetric in the two layers and obeys Onsager reciprocity. To prove this, first consider the case $n_{\text{rms}}^A = 0$ where the active layer is homogeneous. The integral in dn'_A of Eq. (1) may be removed. After cancelling away common factors in the numerator and denominator, we obtain

$$\sigma_D^E = \frac{\int_{-\infty}^{\infty} dn'_P P(n'_P) \cdot \frac{\sigma_D(n_A, n'_P)}{(\sigma_P^E + \sigma_P(n'_P))}}{\int_{-\infty}^{\infty} dn'_P \frac{P(n'_P)}{(\sigma_P^E + \sigma_P(n'_P))}}. \quad (\text{S38})$$

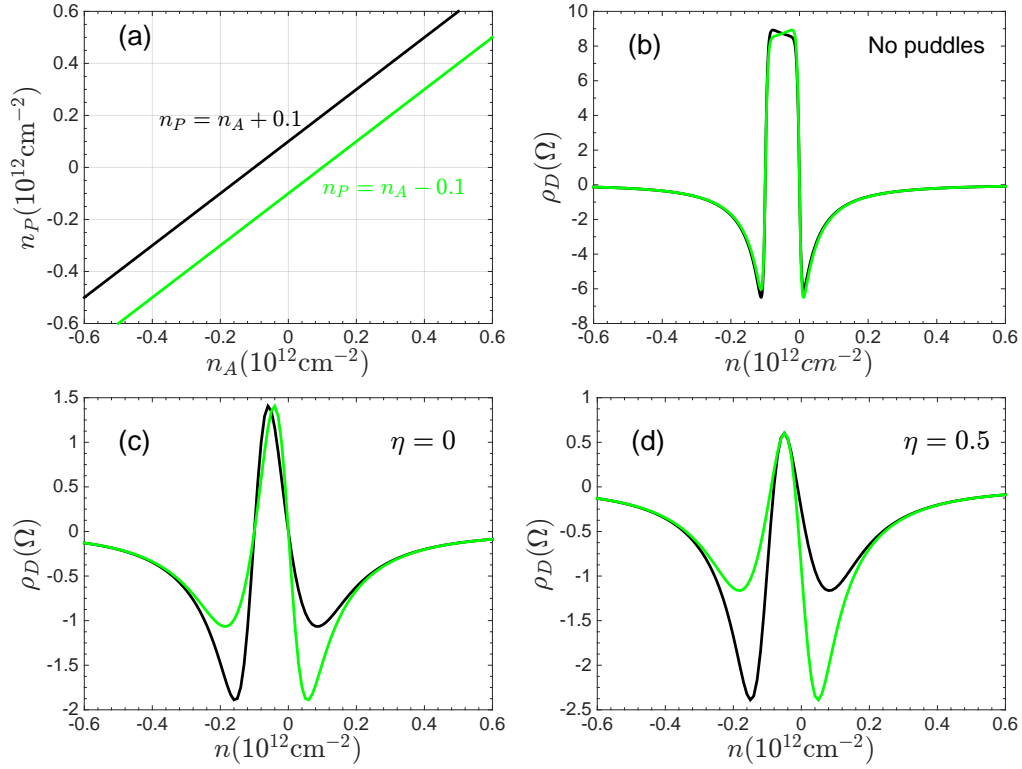


Figure S4. (color online) Predicted experimental signatures of inhomogeneity in the drag resistivity. The behavior of drag resistivity as a function of $n'_A = n$ ($n'_P = n$) along the black (green) line in panel (a) calculated using (b) homogeneous theory (c) uncorrelated charge fluctuations and (d) correlated charge fluctuations. $T = 70\text{K}$, $(d_{\text{imp}}^A, d_{\text{imp}}^P) = (10, 20) \text{ nm}$ in all panels and $(n_{\text{rms}}^A, n_{\text{rms}}^P) = (10, 5) \times 10^{10} \text{cm}^{-2}$ in (c) and (d).

Now if we swap the role of the layers, we will have $n_{\text{rms}}^P = 0$. In this case, Eq. (1) similarly reduces to

$$\sigma_D^E = \frac{\int_{-\infty}^{\infty} dn'_A P(n'_A) \cdot \frac{\sigma_D(n'_A, n_P)}{(\sigma_A^E + \sigma_A(n'_A))}}{\int_{-\infty}^{\infty} dn'_A \frac{P(n'_A)}{(\sigma_A^E + \sigma_A(n'_A))}}. \quad (\text{S39})$$

Because the passive layer of Eq. (S38) and the active layer of Eq. (S39) refer to the same layer due to the swapping of roles, the two equations yield identical σ_D^E . This proves that Onsager reciprocity holds.

We now discuss the rest of the cases where both layers are inhomogeneous, η is an arbitrary number between -1 and 1 excluding zero, and both n_{rms}^A and n_{rms}^P are non-zero. The term in square brackets (i.e. the averaging kernel) in the numerator of Eq. (1) is invariant under the ‘EMT-invariance’ transformation $(n'_A, n'_P, n_A, n_P, n_{\text{rms}}^A, n_{\text{rms}}^P) \rightarrow (-n'_A, -n'_P, -n_A, -n_P, n_{\text{rms}}^A, n_{\text{rms}}^P)$, where the first two arguments refer to the dummy integration variables of Eq. (1), the second and third refer to the average densities and the last two refer to the root mean square density fluctuations in the layers. Notice that the n_{rms} of each layer must be left unchanged so that σ_A^E and σ_P^E retain their original values. The same invariance rule holds true for the term in square brackets in the denominator. Calculation of the EMT drag conductivity may be visualized as averaging these terms over an area in the (n'_A, n'_P) plane determined by $(n_A, n_P, n_{\text{rms}}^A, n_{\text{rms}}^P)$ and η as illustrated in Fig. S3. Swapping the roles of the two layers corresponds mathematically to performing the ‘swap’ transformation $(n'_A, n'_P, n_A, n_P, n_{\text{rms}}^A, n_{\text{rms}}^P) \rightarrow (n'_P, n'_A, n_P, n_A, n_{\text{rms}}^P, n_{\text{rms}}^A)$. The two transformations are generally inequivalent for arbitrary $(n_A, n_P, n_{\text{rms}}^A, n_{\text{rms}}^P)$. This implies that Onsager’s reciprocity will be broken in the majority of situations. For instance, in the case of different density fluctuation amplitudes $n_{\text{rms}}^A \neq n_{\text{rms}}^P$, Onsager reciprocity is certain to break down regardless of the value of the average densities (n_A, n_P) . In Fig. S3, the oval marked ‘original’ represents the area in the (n'_A, n'_P) plane that is averaged over in the calculation of the EMT drag conductivity using Eq. (1). The two correlated layers have average densities (n_A, n_P) and the values of $n_{\text{rms}}^A, n_{\text{rms}}^P$ and η are encoded in the shape of the oval. Swapping the roles of the two layers and calculating the corresponding EMT drag conductivity corresponds to averaging over the oval marked ‘swapped’ which is obtained from the original

one by performing the ‘swap’ transformation on it. The oval marked ‘EMT-invariant’ is obtained by performing the ‘EMT-invariance’ transformation on the original oval. The two calculations will give the same result (i.e. Onsager reciprocity will be obeyed) only if the ‘swapped’ oval exactly overlaps with the ‘EMT-invariant’ oval or the original oval. This can happen only when the original average densities (n_A, n_P) lie along the line $n'_P = -n'_A$ or $n'_P = n'_A$ with $n_{\text{rms}}^A = n_{\text{rms}}^P$. In all of the other cases, the ovals will not overlap and Onsager reciprocity will be violated.

We note in closing this section that at high densities and high temperature, it is possible to prove that Eqs. (1) and (2) reduce to the homogeneous expressions. Hence, at high density and high temperature, Onsager reciprocity will be restored since the homogeneous theory obeys it.

S2. Experimental Signatures of Inhomogeneity

Measurements of Onsager reciprocity violation serve as indicators of the presence of density fluctuations but swapping the layers may not always be experimentally convenient or feasible. We thus propose here an easier method of measuring signatures of the presence of puddles. In this method, one simply swaps the densities of the two layers and compares the drag resistivities measured before and after the swap. In the homogeneous theory, the drag resistivities should be almost the same as shown in Fig. S4(b). In fact, if the energy-independent scattering time were valid or if $d_{\text{imp}} = 0$ in both layers, exchanging densities will cause no change at all to ρ_D . In the presence of charge inhomogeneity however, our EMT numerics shown in Fig. S4(c) and (d) predict that large differences in ρ_D will occur upon swapping the layer densities. This result is relatively insensitive to puddle correlations, as seen from the similarity of the two panels.

HOMOGENEOUS DRAG THEORY

(i) Drag Conductivity Expressions

The dynamically screened interlayer Coulomb interaction is given by

$$V(q, \omega, d) = \frac{V_{12}(q, d)}{\epsilon_D(q, \omega, d)} \quad (\text{S40})$$

where the double-layer dielectric function ϵ_D is given by

$$\epsilon_D(q, \omega, d, T) = (1 - V_{11}(q)\Pi_A(q, \omega, T))(1 - V_{22}(q)\Pi_P(q, \omega, T)) - V_{12}(q)V_{21}(q)\Pi_A(q, \omega, T)\Pi_P(q, \omega, T), \quad (\text{S41})$$

with bare interlayer and intralayer Coulomb potentials $V_{12}(q, d) = V_{21}(q, d) = 2\pi e^2 \exp(-qd)/\kappa q$ and $V_{11}(q) = V_{22}(q) = 2\pi e^2/\kappa q$ respectively, where κ is the dielectric constant of the material encapsulating the graphene sheets. Π_i is the dynamical polarizability of layer i as given in Ref. [34]. At low density, Π_i depends predominantly on temperature and increases with T , whereas at high density, it depends predominantly on charge density n and increases as $\sqrt{|n|}$. The reason for these features is that screening becomes more effective as the total number of carriers (electrons plus holes) increases, and the carrier density at low (high) density depends most sensitively on temperature (charge density).

The nonlinear susceptibility is a response function relating a voltage with the current it induces [26] via

$$\mathbf{i}(\omega) = \int d\mathbf{r}_1 \int d\mathbf{r}_2 \mathbf{\Gamma}(\omega, \mathbf{r}_1, \mathbf{r}_2) V(\mathbf{r}_1) V(\mathbf{r}_2). \quad (\text{S42})$$

The x -component is given by $\Gamma^x(\omega, \mathbf{q}, \mu/k_B T) = \Gamma(\omega, q, \mu/k_B T) \cos(\theta_q)$. It is convenient at this stage to switch to the following dimensionless notation.

$$\tilde{\omega} = \frac{\hbar\omega}{k_B T}, \quad \tilde{q} = \frac{\hbar v_F q}{k_B T}, \quad \tilde{\mu} = \frac{\mu}{k_B T}, \quad \tilde{E} = \frac{E}{k_B T}, \quad z = \frac{2\tilde{E} + \tilde{\omega}}{\tilde{q}}. \quad (\text{S43})$$

With this notation, we follow the approach detailed in Ref. [26] to obtain the expressions

$$\begin{aligned}
\Gamma(\omega, q, \frac{\mu}{k_B T}) &= -\frac{4e}{\hbar v_F} \tilde{\Gamma}(\tilde{\omega}, \tilde{q}, \tilde{\mu}), \\
\tilde{\Gamma}(\tilde{\omega}, \tilde{q}, \tilde{\mu}) &= \frac{1}{4\pi} G(\tilde{\omega}, \tilde{q}, \tilde{\mu}), \tilde{q}, \\
G(\tilde{\omega}, \tilde{q}, \tilde{\mu}) &= \begin{cases} -\frac{1}{2} \int_0^1 dz I(z, \tilde{q}, \tilde{\omega}, \tilde{\mu}) \sqrt{\frac{1-z^2}{\frac{\tilde{\omega}^2}{\tilde{q}^2}-1}} K(z, \tilde{q}, \tilde{\omega}), & |\tilde{\omega}| > \tilde{q}, \\ \frac{1}{2} \int_1^\infty dz I(z, \tilde{q}, \tilde{\omega}, \tilde{\mu}) \sqrt{\frac{z^2-1}{1-\frac{\tilde{\omega}^2}{\tilde{q}^2}}} K(z, \tilde{q}, \tilde{\omega}), & |\tilde{\omega}| < \tilde{q}, \end{cases} \\
I(z, \tilde{q}, \tilde{\omega}, \tilde{\mu}) &= \tanh\left(\frac{z\tilde{q} - \tilde{\omega} - 2\tilde{\mu}}{4}\right) - \tanh\left(\frac{z\tilde{q} + \tilde{\omega} - 2\tilde{\mu}}{4}\right) + \tanh\left(\frac{z\tilde{q} + \tilde{\omega} + 2\tilde{\mu}}{4}\right) - \tanh\left(\frac{z\tilde{q} - \tilde{\omega} + 2\tilde{\mu}}{4}\right), \\
K(z, \tilde{q}, \tilde{\omega}) &= \tilde{\tau} \left(\frac{z\tilde{q} - \tilde{\omega}}{2}\right) \frac{z\tilde{\omega} - \tilde{q}}{z\tilde{q} - \tilde{\omega}} - \tilde{\tau} \left(\frac{-z\tilde{q} - \tilde{\omega}}{2}\right) \frac{z\tilde{\omega} + \tilde{q}}{z\tilde{q} + \tilde{\omega}}, \\
\tilde{\tau}(\tilde{E}) &= \frac{k_B T \tau(E)}{\hbar}.
\end{aligned} \tag{S44}$$

where τ is the transport scattering time. In this paper we assume that electron-charged impurity scattering dominates over all other scattering mechanisms. In this case, τ is given by

$$\frac{1}{\tau(E)} = \frac{4\pi n_{\text{imp}}}{\hbar} \int \frac{d^2 k'}{(2\pi)^2} \left| \frac{V_{\text{imp}}(q)}{\epsilon_s(q)} \right|^2 \frac{1 - \cos^2(\theta_{\mathbf{k}, \mathbf{k}'})}{4} \delta(E_k - E_{k'}), \tag{S45}$$

where $q = |\mathbf{k} - \mathbf{k}'|$ and $\theta_{\mathbf{k}, \mathbf{k}'}$ is the angle between the initial and final wave vectors \mathbf{k} and \mathbf{k}' in a scattering event. n_{imp} is the areal concentration of charged impurities in the assumed impurity plane near the graphene sheet. $V_{\text{imp}}(q) = 2\pi e^2/(\kappa q) \exp(-qd_{\text{imp}})$ where d_{imp} is the distance between the graphene sheet and the charged impurities which are assumed to lie in a single plane. We shall assume for now that $d_{\text{imp}} = 0$ in both layers unless otherwise specified. We also assume equal impurity concentrations in both layers so that $n_{\text{imp}}^A = n_{\text{imp}}^P \equiv n_{\text{imp}}$. The general case is recovered from all the expressions by simply replacing n_{imp}^2 with $n_{\text{imp}}^A n_{\text{imp}}^P$.

The single layer dielectric function ϵ_s is given by

$$\epsilon_s(q) = 1 - \Pi(q, T) V_{11}(q), \tag{S46}$$

where $\Pi(q, T)$ is the static polarizability of graphene. As pointed out in Ref. [26], the nonlinear susceptibility contains a logarithmic divergence along the line $\tilde{q} = \tilde{\omega}$ so long as τ has an E -dependence. In this work, we prevent σ_D from diverging in calculations by using the dynamical polarizability of Ref. [34] in the dielectric function. This introduces a divergence in the denominator of σ_D which cancels that in the numerator, leaving behind a finite and well-defined quantity. We shall assume the value of the graphene ‘fine structure constant’ is $r_s = 0.568$, corresponding to an estimated value of $\kappa = 3.5$ for hexagonal boron nitride. The above expressions constitute all the ingredients one needs to calculate the drag conductivity.

(ii) $F(r_s, \frac{k_B T d}{\hbar v_F})$ for low density σ_D Asymptote

Here we plot the numerical coefficient $F(r_s, \frac{k_B T d}{\hbar v_F})$ which features in the low-density drag conductivity asymptote Eq. (4) as a function of $k_B T d / \hbar v_F$ for different values of r_s .

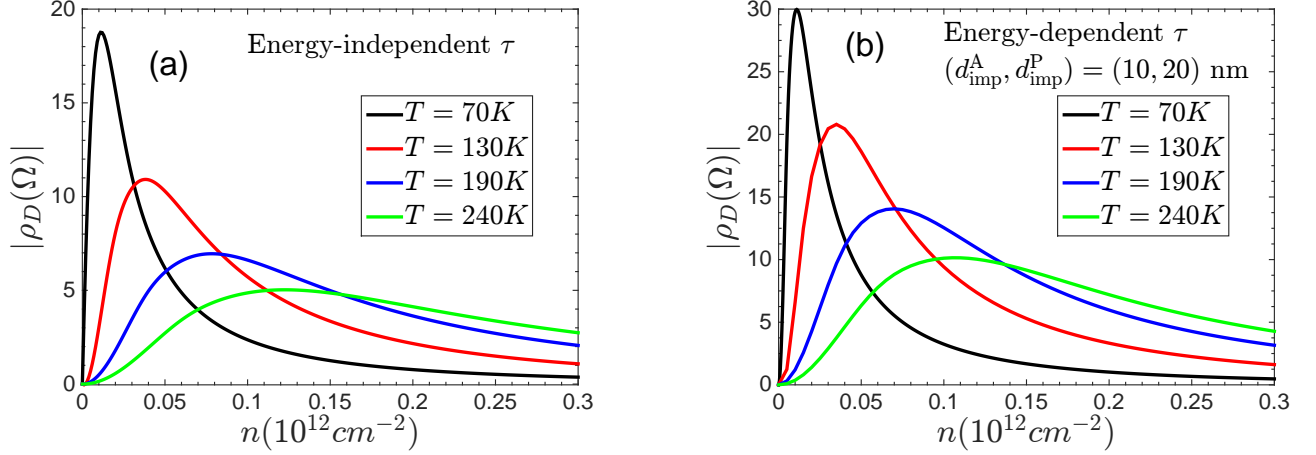


Figure S6. (color online) Quantitative effect of transport scattering time energy-dependence on $|\rho_D|$. $|\rho_D|$ calculated as a function of charge density along lines of identically matched density $n_A = n_P = n$ at different temperatures for (a) Energy-independent scattering time and (b) Energy-dependent (electron-impurity) scattering time, where one graphene sheet is nearer the impurity plane. Interlayer spacing $d = 9$ nm.

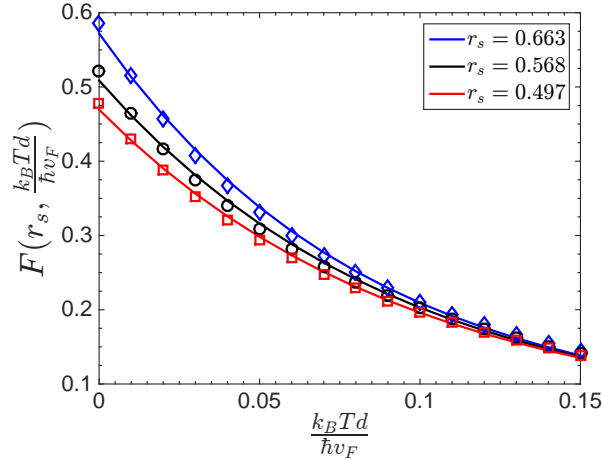


Figure S5. (color online) Values of the numerical constant $F(r_s, \tilde{d})$ as a function of $\tilde{d} \equiv k_B T d / \hbar v_F$ for different values of r_s . Data points represent numerical evaluations of $F(r_s, \tilde{d})$ while lines represent fits of the form in Eq. 5. For $r_s = (0.663, 0.568, 0.497)$, we have $a = (0.52, 0.46, 0.42)$ $b = (11.95, 10.25, 10.34)$ $c = (0.05, 0.04, 0.05)$ respectively. Note that these values of r_s correspond to dielectric constants of 3.5, 4 and 3 respectively. The dielectric constants of common spacer materials such as hBN, SiO₂ and Al₂O₃ all fall between 3 and 4.

(iii) Energy-Independent vs Energy-dependent Scattering Time

Figures. S6(a) and (b) show $|\rho_D|$ calculated as a function of charge density at different temperatures, assuming energy-independent and energy-dependent scattering times (see Eq. (S45)) respectively. The drag peaks at finite density differ by up to a factor of about two due to the difference in scattering time. We emphasize that these calculations are for the completely homogeneous case. They show that the widely accepted practice of using energy-independent scattering times may be problematic. Note that the curves in Fig. S6(a) closely resemble those in Fig. 1(d) in which $d_{\text{imp}}^A = d_{\text{imp}}^P = 0$. This seems to suggest that when the two layers are identical, the energy-independent

scattering time is a good approximation.

(iv) Relation between Charge Density and Chemical Potential

Here we review the one-to-one correspondence between charge density and chemical potential given a fixed temperature. The charge density is defined as $n = n_e - n_h$ where n_e and n_h refer to the electron and hole densities respectively. These densities are obtained from the chemical potential μ and temperature T via

$$n_{e,h} = -\frac{2}{\pi} \left(\frac{k_B T}{\hbar v_F} \right)^2 \text{Li}_2 \left[-\exp \left(\pm \frac{\mu}{k_B T} \right) \right], \quad (\text{S47})$$

where Li_2 refers to the dilogarithm function. This equation allows us to find the charge density given the chemical potential and temperature.

We can also find the chemical potential given the charge density and temperature. This is done by noting that $E_F = \hbar v_F \sqrt{\pi |n|} \text{sign}(n)$ and using the relation

$$\frac{\mu}{E_F} = F_\mu \left(\frac{k_B T}{|E_F|} \right), \quad (\text{S48})$$

where

$$F_\mu(x) = \bar{g}(x) \left(1 - \frac{\pi^2 x^2}{6} \right) + g(x)/[4 \log(2)x], \quad (\text{S49})$$

with $g(x) = (1 + \text{Erf}[10(x - 0.5)])/2$ and $\bar{g}(x) = \text{Erfc}[10(x - 0.5)]/2$. Eqs (S47) and (S48) allow one to trivially convert a function of charge density to a function of chemical potential and vice versa.

(v) Monolayer Conductivity

The monolayer conductivity is given by

$$\sigma(\mu) = \frac{e^2}{2} \int dE D(E) v_F^2 \tau(E) \left(-\frac{\partial f(E, \mu, T)}{\partial E} \right), \quad (\text{S50})$$

where $i = A, P$, $D(E) = 2|E|/(\pi \hbar^2 v_F^2)$ is the density of states and $f(E, \mu, T)$ the Fermi function, given by

$$f(E, \mu, T) = \frac{1}{\exp(\frac{E - \mu}{k_B T}) + 1}, \quad (\text{S51})$$

and $\tau(E)$ is given in Eq. (S45). We plot monolayer conductivity as a function of charge density in Fig. S7. The monolayer conductivity at low density goes as the square of temperature and is given by

$$\sigma = C_0 \frac{(k_B T)^2}{\hbar^2 v_F^2 n_{\text{imp}}} \frac{e^2}{\hbar}, \quad (\text{S52})$$

where $C_0 = 3.18$ for $r_s = 0.568$. At high density, σ goes linearly in n (quadratically in μ) with weak metallic corrections. Modifying this equation to take into account our different r_s of 0.568, we obtain

$$\sigma(T) = \sigma_0 \left[1 - \frac{\pi^2}{3} 0.25 \left(\frac{T}{T_F} \right)^2 \right], \quad (\text{S53})$$

where σ_0 is the zero temperature conductivity obtained by taking the $T = 0$ limit in Eq. (S50). Note also that the typical value of σ is about three orders of magnitude larger than that of σ_D (cf. Fig. 1(b)), confirming that Eq. (10) is valid.

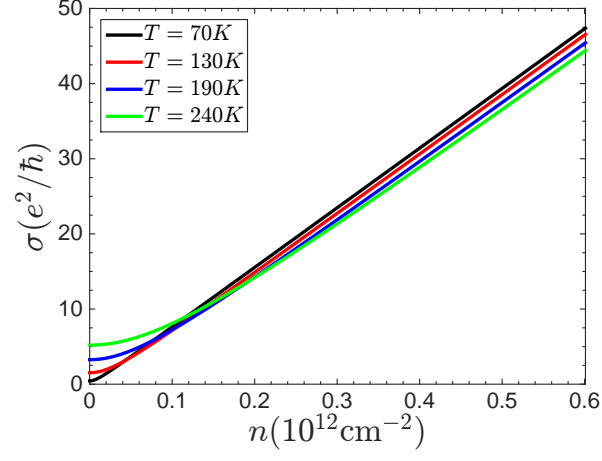


Figure S7. (color online) The monolayer conductivity as a function of charge density n at different temperatures as given by Eq. (S50). Note that σ is an even function of n . $n_{\text{imp}} = 5 \times 10^{10} \text{ cm}^{-2}$.